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Field Campaign Name (50 characters)
Aerosol Lifecycle IOP at BNL

AMS Site Designation
AMF

Scientific Focus
Succinct statement of underlying hypothesis or technical goal for proposed research.

Conduct intensive aerosol observations in a region exposed to anthropogenic, biogenic, and marine emissions with atmospheric processing times depending on air mass trajectories and time of day. Take advantage of new instruments in the MAOS (e.g., SP2, HR-PTRMS, ACSM, Trace Gas Suite, PASS-3, Aethelometer, UHSAS).

There are three main foci of the planned IOP:
1. Aerosol light absorption: How does the aerosol mass absorption coefficient (absorption per unit mass of BC) vary with atmospheric processing? Do observations agree with a shell-core model?
2. Secondary organic aerosol (SOA): How does the amount and formation rate of SOA vary with atmospheric processing and sources? Can heretofore unavailable measurements of oxygenated VOCs (from high resolution PTR-MS) explain the "excess" SOA observed in other locations?
3. Aerosol as CCN: What is the effect of different organic components on CCN formation?

Relevancy to ARM Mission
Please refer to our mission statement.

It is recognized that "Improving the fidelity and predictive capability of global climate models requires better understanding of a multitude of fundamental aerosol and cloud lifecycle processes" (ASR, 2010). The proposed IOP address aerosol lifecycle elements that are poorly represented in models, yet important on local and global scales to determining radiative forcing.

A continuing issue is the quantification of the black carbon (BC) contribution to aerosol radiative forcing. To date, this has proved difficult because the optical properties of BC are closely tied to its morphology, which, in turn, is dependent upon how it is generated and its aging behavior in the atmosphere. While laboratory efforts can help elucidate isolated aspects of this issue (e.g., Cross, et al. 2010), field measurements are necessary to document actual behavior in the atmosphere and to help guide the direction of future laboratory experiments. A challenge is to relate BC emissions to atmospheric optical and radiative effects in ways useable in global models.

Organic compounds contribute approximately 50% to the global burden of anthropogenic fine mode aerosols (Zhang et al., 2007). This fraction is under-predicted in models, sometimes by as much as an order of magnitude. There are downstream effects of organic aerosol upon lifetime, light scattering and absorption, and cloud condensation nuclei (CCN) forming potential that ultimately lead to radiative impacts. The challenge is to identify the sources of secondary organic aerosols, so that they can be incorporated in emission based climate models.

Currently, aerosol indirect effects represent the largest uncertainties in anthropogenic radiative forcing derived from global climate model simulations. The large uncertainties in indirect effects are in part due to crude representations of aerosol CCN properties and the subsequent treatment of cloud activation in global models. Whereas the cloud activation properties of inorganic species typically found in atmospheric aerosols are well understood, the effects of organics species are poorly understood, yet they can contribute significantly to, or sometimes dominate, the total fine aerosol mass. A challenge is to quantitatively define the extent to which details of aerosol composition are important, and thereby need to be represented in global models. Further, to develop parameterizations for global models that represent effects of the hundreds or even thousands of organic species actually in ambient aerosol.

Atmospheric System Research (ASR) Science and Program Plan, Jan., 2010, U.S. DOE, Office of Science

Cross, E., S., T. B. Onasch, A. A., W. Wrobel, J. G. Slowik, J. Olfert, D. A. Lack, P. Massoli, C. D. Cappa, J. Schwarz, R. Spackman, D. W. Fahey, A. J. Sedlacek, A. Trimborn, J. T. Jayne, A. Freedman, L. R. Williams, N. L. Ng, C. Mazzoleni, M. Dubey, B. Brem, G. Kok, R. Subramanian, S. Freitag, A. Clark, D. T. Thornhill, L. Marr, C. E. Kolb, D. R. Worsnop, and P. Davidovits, (2010) Soot Particle Studies – Instrument Inter-Comparison – Project overview *(accepted for publication, AS&T)*

Zhang, Q., and coauthors, 2007, Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere Mid-latitudes, *Geophys. Res. Lett.*, 34, L13801, doi:10.1029/2007GL029979.

Description

The description should include the general work plan and the experimental methods and nature of the measurements to be made. It should include information about the spatial

and temporal nature of the data collection, methods for data analysis, and expected final product (e.g., a final report, a publication, etc.). All data from ARM field campaigns must be delivered to the ARM Data Archive within six months of completion of the field campaign.

The proposed field campaign is an aerosol IOP to occur over an 8 week period during the summer of 2011. Measurements will be made on site at the BNL "Met. Field", which is a large open area that has been used over many decades for meteorological monitoring and occasionally for chemical measurements. The site has electric power and internet connectivity. From past measurements at the surface and from the G-1 aircraft (Kleinman et al., 2000), we expect that different types of emissions can dominate depending on meteorology. A ground site at BNL can receive urban emission dominated air from the west and southwest, and biogenic emission dominated air from the north and northeast with a distance from the source regions corresponding to atmospheric transport time of hours to days. Absent strong synoptic forcing, a sea breeze develops in the afternoon. Over an 8 week period, a few haze events (pollution alerts) can be expected and there is perhaps a 50% chance of catching an intense but distant biomass burning event.

Studies of BC, SOA, and CCN are described below. A key component of these three focus areas is that aerosol properties will be determined as function of atmospheric processing and chemical conditions or source type. Sources of aerosols and their precursors will be determined from chemical tracers (e.g., CO, CH_3CN, other VOCs, and SO_2). Atmospheric processing will determined from back trajectories and photochemical age.

1. Optical effects of BC

BC mass concentration from the SP2 will be combined with light absorption measurements (PSAP, PASS, and PTI) to determine a mass absorption coefficient. Coating thickness will be determined from the SP2 using its luminescence and scattering channels (thin/thick coating) and by comparing aerosol size distributions with and without a thermal denuder. Aerosol composition from the thermal denuder and from the AMS and PILS will provide information on the coating material. Theory and observations will be compared.

2 SOA Formation.

Total OA concentration, along with that of NH_4, SO_4 and NO_3, will be determined using an Aerodyne Aerosol Mass Spectrometer (AMS). Concentrations of SOA will be approximated by oxygenated-OA (OOA) evaluated from factor analysis of the AMS data (such as the PMF). CO is a good tracer for urban emissions and will be used to assess extent of dilution during transport. Black carbon (by SP2) will be used to estimate primary organic aerosol (POA) using a known emission ratio at the source, which can be checked against the POA taken as the hydrocarbon-like OA (HOA) estimated also from factor analysis.

Volatile and oxygenated organic compounds will be quantified using a high resolution Proton Transfer Reaction – Mass Spectrometer (HR-PTRMS), which provides source information based on relative abundance of anthropogenic and biogenic compounds (e.g., benzene vs. isoprene/methyl vinyl ketone) as well as photochemical age (e.g. benzene/toluene ratio). The distinguishing feature of this study is high mass resolution so that oxygenated VOCs can be differentiated from hydrocarbons of nearly the same mass. By following the oxygen content, the HR-PTRMS offers a measure of the extent of VOC oxidation in an air mass. We will investigate the dependence of the extent and rate of SOA formation upon VOC oxidation. Theories that predict that "excess" SOA is due to the condensation of oxygenated VOCs will be tested and perhaps key oxygenated species involved in SOA formation will be revealed.

Cloud processing of organic vapor as a pathway for SOA formation will be investigated by comparing SOA formation and changes in gas phase organics on days with and without clouds.

3 CCN

The cloud activation properties of major aerosol organic classes will be determined from simultaneous size-resolved measurements of cloud condensation nuclei (CCN) spectra, mixing state (HTDMA in MAOS-A), and particle composition. These measured CCN properties of organic classes can be conveniently incorporated into parameterizations for improved representation of aerosol-cloud interaction in global climate models. Based on measured aerosol size distribution and composition, CCN spectra will be calculated using various simplified representations of aerosol composition, and compared to direct measurements.

Data taken during the proposed experiments will be added to the ASR archive and made public in accordance to ASR policy. Results will be reported in the peer reviewed literature.

Kleinman, L. I., Daum, P. H., Imre, D. G., Lee, J. H., Lee, Y.-N., Nunnermacker, L. J., Springston, S. R., Weinstein-Lloyd, J., and Newman, L., 2000, Ozone production in the New York City Urban Plume. J. Geophys. Res. 105, 14,495-14,511.

ARM Resources Needed

What on-site ARM instruments are to be used in the campaign? Will additional instruments be brought to the site? Over what period of time will investigators and technicians be on site (a Site Access Request must be completed for each site visitor)? Are ARM staff technicians needed to establish and maintain additional instruments? What are the data transfer requirements? **For the instrument development, include expected labor, procurement cost, and/or number of test flight hours (if requested)**.

We are requesting MAOS-A and MAOS-C, with the latter having higher priority. BNL will supply instruments for size resolved CCN measurements. The BNL photothermal

interferometer (PTI) will be used to measure aerosol absorption alongside the PAS and PSAP in MAOS-C. In addition to the ACSM of MAOS, an Aerodyne AMS will be required for measurement of aerosol composition, including organic mass spectra related to source and atmospheric processing. A c-ToF-AMS with unit mass resolution is available at BNL. We are pursuing collaborations to bring a high resolution AMS to BNL.

Aircraft
No

Additional Information

While the DOE guidelines suggest a deployment time of at least 6-10 months for the ARM Mobile Facilities and state that preference will be given for deployments in which the AMF is embedded in a large field campaign, there are in this instance good reasons for conducting a more focused campaign of short duration:

1. The science questions posed here can be addressed over a time period that covers several synoptic cycles, circa one week apiece.

2. Questions posed can be addressed without multiple platforms.

3. MAOS-C contains a suite of instruments that have been used by the ASP community but are new to the ARM community. There are legitimate questions as to the feasibility of operating MAOS-C for extended periods of time without cadre of support personal that are impractical for long deployments.

4. It is likely that the summer of 2011 will be the first opportunity to use MAOS-A and C, and a short deployment would be very valuable in finding and fixing problems before these units are set on their way.